

[From the Author.]

## III.

## THE VERMICULITES:

## THEIR CRYSTALLOGRAPHIC AND CHEMICAL RELATIONS TO THE MICAS;

## TOGETHER WITH A DISCUSSION OF THE CAUSE OF THE VARIATION OF THE OPTICAL ANGLE IN THESE MINERALS.

BY JOSIAH P. COOKE, JR.,

Erving Professor of Chemistry and Mineralogy in Harvard College.

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*Introduction.* — In the American Journal of Science (VII. 55, 1824), T. H. Webb described a mineral from Millbury, near Worcester, Mass., which has since been a mineralogical curiosity on account of its singular reaction when heated. The mineral consists of "small foliated scales distributed through a steatitic base." "When heated, it exfoliates prodigiously, the scales opening out into long worm-like threads made up of the separate folia. Exfoliation commences at 500° to 600° Fahr., and takes place with so much force as often to break the test-tube in which the mineral may be confined. Before the blowpipe it fuses at 3.5 to a grayish-black glass." It was named by Webb, as he says, "from the Latin *vermiculor*, 'I breed worms.'" The hardness of the mineral is 1-2, the specific gravity 2.756, the lustre talcose, and the color grayish, somewhat brownish. It was analyzed by Crossley, who "separated with great care from the base the scaly mineral, which is the true vermiculite," and his results were as follows:—

	Oxygen Ratio.				Oxygen Ratio.			
	1.		True	App.	2.		True	App.
Silica . . .	35.74	19.06	19.06	11	35.74	19.06	19.06	2
Alumina . .	16.42	7.65	7.65	4	16.42	7.65		
Ferric Oxide .					11.13	3.34	10.99	1
Ferrous Oxide	10.02	2.23						
Magnesia . .	27.44	10.98	13.21	7	27.44	10.98	10.98	1
Water . . .	10.30	9.16	9.16	5	10.30	9.16	9.16	1
	<u>99.92</u>				<u>101.03</u>			

The results of analysis in column 1, and the portions of the description of the mineral in quotation-marks, above, have been taken from "Dana's System of Mineralogy," fifth edition, page 493; and the atomic ratio which is there deduced is,

$$\overset{\text{IV}}{\text{Si}} : \overset{\text{VI}}{\text{R}} : \overset{\text{II}}{\text{R}} : \overset{\text{II}}{\text{H}} = 11 : 4 : 7 : 5.$$

In this analysis, however, Crossley could not have determined the state of the iron, which, in the specimen I have examined, is almost wholly in the ferric condition. If now we assume that the whole of the iron belongs with the sesquioxide radicals, the analysis would appear as in column 2, and the atomic ratio is then seen to be 2 : 1 : 1 : 1, which is undoubtedly the correct result.

In the year 1851, Mr. W. W. Jefferis, of West Chester, Pa., discovered at the ripidolite locality near that town a peculiar micaceous mineral which exfoliates like the Millbury vermiculite, but which, instead of occurring in small foliæ, is found in large hexagonal plates. This mineral was analyzed by Professor Brush; and although at first referred by him, "with a query," to vermiculite (*Am. Jour. Sci.*, II. xxxi. 369, 1861), was subsequently described as a new species (*Am. Jour. Sci.*, II. xli. 248, 1866), and named Jefferisite.

Several years later, Mr. John Hall, now of Philadelphia, sent to me for examination some rough six-sided prisms of a micaceous mineral, which he had discovered at East Nottingham, in Chester County, Pa. This mineral also exfoliates when heated. It is a new species, and I have named it, after the discoverer, Hallite.

A year since I received from Colonel C. W. Jenks, in connection with other minerals from his corundum mine on the Culsagee River, in Macon County, N. C., a specimen of still another micaceous mineral having the same remarkable pyrognostic properties. It proved to be the best defined of any of this class of minerals which I had examined, and I shall designate it as Culsageeite.

Besides the above, there have been found several other micaceous minerals whose pyrognostic and crystallographic characters indicate that they belong to the same family, but which have not yet been investigated.

The remarkable exfoliation and great apparent increase of volume which the class of mineral under consideration undergo when heated are analogous to the well-known phenomena presented in the dehydration of alum, borax, and other crystalline salts, when heated in a similar way; and it will be one object of this paper to show that the effect is due to the same cause, — namely, to the escaping of what we call water



of crystallization. I also expect to show that the several minerals referred to above are members of a family of hydrous silicates closely allied and parallel to the well-known family of anhydrous silicates called the micas, and that their molecules differ from those of the magnesian micas chiefly in containing a definite number of molecules of water; that is, water of crystallization. I shall call this family of minerals "the vermiculites," using the original name, as "mica" is now employed, to designate a class; and I shall call the three species (or varieties?) of this family Jefferisite, Culsageite, and Hallite; which correspond, as I expect to show, to the two varieties of Biotite and to Phlogopite respectively. It will appear that the original vermiculite has the same composition as the variety from the Culsagee mine. Finally, attention will also be asked to some unexpected discoveries to which the optical examination of these minerals has led.

*Water of Crystallization and Water of Constitution.*—In the salts of the stronger acids, the distinction between water of constitution (basic water, as it is sometimes called) and water of crystallization is, as a rule, well marked. Water of crystallization generally escapes at a comparatively low temperature, and the loss is attended with a complete disintegration of the crystals, which usually fall to powder. Water of constitution is held more firmly, and the salt must be heated to a much higher temperature before it is expelled. Moreover, although the loss of basic water may greatly alter the external aspect of the body, yet there is not the same complete breaking up of the structure as before. To illustrate these points, it is only necessary to refer to the well-known reactions of the common rhombic sodic phosphate when heated.

It is generally believed that the water of crystallization exists as such in the salt, the molecules of water forming a part of the structure of the crystals; and the facts to be presented in this paper tend to support this view. But our modern theories assume that the so-called water of constitution, which may be also driven off when the salt is sufficiently heated, is formed under the influence of the heat from the atoms of hydroxyl  $H-O-$ , which are united to the acid radical of the compound. But in every simple salt, whatever may be our theories in regard to the mode of combination, the weight of the water of constitution, as well as that of the water of crystallization, must bear to the weight of the salt a definite proportion, which can easily be calculated from the symbol; and the ratio between the amount of hydrogen and the amounts of the other radicals must be that of the atomic weights of these radicals or of their multiples. In any given class of salts,

moreover, in which the ratio between the atomicity of the basic and acid radicals has a constant value, all hydrogen, which represents water of constitution, will supplement the other basic radicals, and added to them will complete the relative amount of basic radical which the given class of salts require; while the hydrogen, which represents water of crystallization, will be in excess of that amount.

In mineral silicates these relations are complicated by the phenomena of isomorphous replacements, and, although there may be some question in regard to the replacing capacity of hydrogen, we cannot expect the same constancy in the amount of water of constitution, which these minerals contain, as in the case of simple salts. The amount of water of crystallization, however, must be as invariable in one case as in the other, and the hydrogen must bear the same relations to the atomic ratio of the compound. Thus, if the mineral is an orthosilicate in which the atomic ratio between the *sum of the basic radicals* and the *silicon* is 1:1, all hydrogen in excess of the amount which this ratio requires must represent water of crystallization, while all required to complete the ratio must represent water of constitution; and we thus have a means of distinguishing between these two states of combination where the class of compounds to which the mineral belongs is known.

Now, it is true of each of the minerals we have distinguished among the vermiculites, —

*First.* That the water is driven off at a temperature below a red heat.

*Secondly.* That the loss of the water is attended with a complete disintegration of the mineral.

*Thirdly.* That the amount of the basic radicals, exclusive of the hydrogen, is sufficient to saturate the silicon, and that the amount of hydrogen is wholly in excess of the amount which the atomic ratio 1:1 requires.

Could it be proved that the vermiculites are orthosilicates, the last of the three facts just stated would be alone sufficient to establish the correctness of our conclusion. Unfortunately such absolute proof cannot be obtained; and we only claim that the crystallographic and chemical relations of the vermiculites to each other, and to the magnesian micas, gives a very high degree of probability to our theory that they are orthosilicates.

From these facts we have concluded that the water which enters into the composition of the vermiculites is water of crystallization. For the evidence of the facts, we refer to the descriptions of the several species given below. But, further, in order to justify our conclusion, we pro-



pose to bring into comparison with the vermiculites a class of hydrous micas from which the water obtained is clearly water of constitution, and this class of minerals we shall first describe.

*Damourites*. — Delesse originally gave this name to a hydrous mica which occurs in fine scales in Pontivy in Brittany. Since then, micas of similar composition have been observed in several countries, and shown to be not unusual constituents of granitic rocks.\* Among these we may distinguish several varieties, (or species?) marked by slight differences of composition and optical characters. But we would propose to give the name Damourite to the whole class, distinguishing the varieties by separate names only so far as may be thought necessary. Under the family of Damourites, then, we class all unisilicate micas, which are chiefly silicates of aluminum and potassium, but in which a portion of the alkaline radical is replaced by hydrogen.

*Sterlingite*. — A remarkable mineral of the Damourite type is found at Sterling, Mass., associated with spodumene, in a vein of a large boulder rock. This mineral, for the sake of distinction, I have called Sterlingite; but it does not differ from the original Damourite of Delesse except in the value of the optical angle. We give in parallel columns the characters of the two minerals: —

## STERLINGITE.

Mica-like in structure. An aggregate of flexible and unelastic laminae, frequently an inch in diameter.

Lustre, pearly.

Color, yellow or yellowish-white.

H = 2 — 3.

Sp. gr. at 28°.

1st ex. gave 2.832.

2d „ „ 2.828.

Cleavage basal, highly perfect, as in mica. Jointed parallel to the sides of a rhomb, having an angle of 120°, and also parallel to the shorter diagonal of the same. The markings of these joints, or cleavages, visible on the surfaces of the laminae.

Double refraction strongly negative.

Biaxial with plane of axes parallel to shorter diagonal. Divergence, about 70°. Dispersion of axes very small.

Before the blowpipe fuses on the edges with potash flame.

## DAMOURITE.

Mica-like in structure. An aggregate of fine scales.

Lustre, pearly.

Color, yellow or yellowish-white.

H = 2 — 3.

Sp. gr. = 2.792.

Double refraction negative. Biaxial divergence, 10 to 12 degrees.

\* See the papers of Professor Houghton, cited below.

ANALYSIS.					
STERLINGITE.			DAMOURITE.		
		Oxygen.	1.	2.	Oxygen.
$SiO_2$	43.87	23.40	45.22	43.41	23.15
$Al_2O_3$	36.45	17.00	37.85	35.17	16.39
$Fe_2O_3$	3.36	1.00		4.62	1.39
$MgO$				1.40	0.56
$K_2O$	10.86	1.84	11.20	10.90	1.85
$H_2O$	5.19	4.61	5.25	4.50	4.00
	99.73		99.52	100.00	

Of the two analyses of Damourite, No. 1 is by Delesse, of the mineral found in the gangue of cyanite, at Pontivy in Brittany, and No. 2 is by Igelström, of the similar mineral, found at Horrsjöberg, Wermland. The analysis of Sterlingite was made by Mr. C. E. Munroe, Assistant in the Chemical Laboratory of Harvard College, and the above numbers have been abundantly confirmed in repeated analyses by various students in the same laboratory. The alkalis were treated by Smith's process, and the potassium was weighed as  $PtK_2Cl_6$ . This value, compared with the total weight of the alkaline chlorides, and that of the chlorine also determined, showed that the alkali in the mineral was almost wholly potash, although the presence of lithium and sodium was plainly indicated by the spectroscope. The water was determined by igniting the mineral in coarse powder, previously dried at  $100^\circ C$ . Even after ignition, the finely pulverized mineral is only partially decomposed by hydrochloric acid, and in the above analysis it was decomposed by fusion with sodic carbonate. The usual tests failed to indicate the presence of fluorine.

Regarding the water as basic, and as forming a part of the protoxides, the atomic ratio in Sterlingite, between the silicon, the sesquioxide radicals, and the protoxide radicals, is

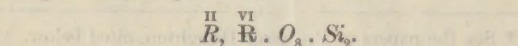
$$\overset{IV}{Si} : \overset{VI}{R} : \overset{II}{R} = 23.40 : 18 : 6.45, \text{ or nearly } 4 : 3 : 1.$$

The deviation from the simple ratio will not appear so great, as seems at first sight, if it is noticed that a difference of one-half per cent in the amount of water would make the ratio almost exact.

The corresponding ratio in the Damourite from Wermland is

$$23.15 : 17.78 : 6.41, \text{ or, as before, nearly } 4 : 3 : 1.$$

These ratios point to the general formula:—





The Damourite and Sterlingite are types of a very large class of hydrous micas, which in many places are widely distributed through the granitic rocks. This class of minerals has been especially investigated by Professor Haughton, of Dublin, whose papers may be found in *Phil. Mag.*, IV. ix. 272, and *Q. J. G. Soc.*, xviii. 414, also xx. 280. We cite here a few of his analyses, selected from those given by Professor Dana, — pages 310 and 311, of the fifth edition of his “*System of Mineralogy*,” — in further illustration of the subject we are discussing:

## DUBLIN CO. — HAUGHTON. Optical Angle, 53°.

Si	Al	Fe	Mg	Ca	Na	K	H
43.47	31.42	4.79	1.13	1.38	1.44	10.71	5.43 = 99.77
23.18	14.64	1.44	0.45	0.39	0.37	1.82	4.83
23.18	16.09		3.03				4.83
23.18	16.09		7.86				
3.	2.		1.				

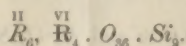
## MT. LEINSTER. — HAUGHTON. Optical Angle, 72°.

Si	Al	Fe	Mg		K	H
44.64	30.18	6.35	0.72		12.40	5.32 = 99.61
23.81	14.07	1.90	0.29		2.11	4.73
23.81	15.97		2.4			4.73
23.81	15.97		7.13			
3.	2.		1.			

## GLEN DALOUGH. — HAUGHTON. Optical Angle, 70°.

Si	Al	Fe	Mg	Ca	Na	K	H
44.71	31.13	4.69	0.90	1.09	1.27	9.91	6.22 = 99.92
23.84	14.51	1.41	0.36	0.31	0.33	1.68	5.53
23.84	15.91		2.68				5.53
23.84	15.91		8.21				
3.	2.		1.				

In these micas again, if we regard the water as basic, we obtain a nearly constant ratio, but differing from that of Damourite in the relation of the two basic radicals. The general symbol of the last would be:—



There appear to be, therefore, these two distinct types of hydrous micas related to the species Muscovite, all rich in alumina and alkali, destitute, or only containing very small amounts, of magnesia, and having a wide optical angle.

The anhydrous Muscovites have not been investigated nearly as fully as the hydrous varieties; and I can find no analyses of any of the beautiful specimens from our American localities. I cite here, for the sake of comparison, an analysis of the Fuchsite, from the Zillerthal, by Schafhäütl (No. 1), and one of a mica, from Fahlun, by H. Rose (No. 2):—

	Si	Al	Fe	Cr	Mg	Ca	Na	K	F
(1)	47.95	34.45	1.80	3.95	0.71	0.59	0.37	10.75	0.35 = 100.92
	25.57	16.05	.54	1.24	.28	0.17	0.09	1.83	
	25.57	17.83				2.37			

	Si	Al	Fe	Mg	K	H	F
(2)	46.22	34.52	6.04	2.11	8.22	0.98	1.03
	24.65	16.08	1.81	0.84	1.39	0.87	
	24.65	17.89			3.10		

If now we should add to (1) 6.93 per cent water, and to (2) 3.42 per cent, we should obtain the ratios:—

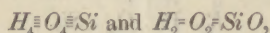
$$(1) \quad 25.57 : 17.83 : 8.52, \text{ or approx. } 3 : 2 : 1,$$

as in the hydrous micas analyzed by Haughton.

$$(2) \quad 24.65 : 17.89 : 6.16, \text{ or approx. } 4 : 3 : 1,$$

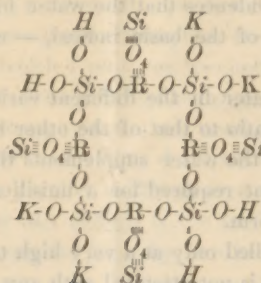
as in Damourite and Sterlingite.

There would seem, then, to be two definite varieties of hydrous micas of the Muscovite family, distinguished by the atomic ratios 3 : 2 : 1 and 4 : 3 : 1. Corresponding to these, it is probable that there are two varieties of anhydrous mica, containing an excess of  $SiO_2$ , which, by the addition of sufficient water to saturate the excess of the acid radical, are reduced to one or the other of the two normal types. In a previous paper (Am. Jour. Sci., II. xii. 217, 1867) the author suggested the idea that the excess of silica in this class of micas might result from a mixture of two isomorphous species corresponding to the two hydrates:—

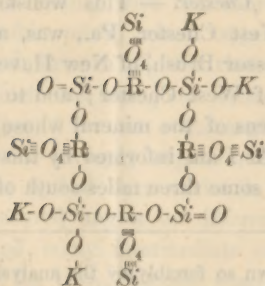




and he there described a mica whose atomic ratio was very closely that of the second type, and which he called Cryophyllite. Now, the simplest theory of the relation of the hydrous to the anhydrous Muscovites would seem to be that, while in the molecules of the anhydrous micas a portion of the silicon atoms (the number varying in the different varieties) are in the condition of the first anhydride ( $H_2=O_2=SiO$ ), the hydrous micas contain sufficient basic hydrogen to bring all the silicon atoms into the condition of the normal hydrate ( $H_4=O_4=Si$ ). The two graphic formulæ which follow indicate, more clearly than any other language can, the relations we have attempted to describe. In these formulæ R stands for the double atom in the radical of the sesquioxides, having the quantivalence of six:—



*Hydrous Muscovite.*



*Anhydrous Muscovite.*

The new mineral Sterlingite, whose examination has been the occasion of this discussion, is remarkable as being a very well-defined example of a hydrous mica occurring in large crystals, and exhibiting very marked characters. It does not materially differ in composition from Damourite, and it also agrees with the specimens of this mineral so closely in other physical qualities that we cannot regard the small

optical angle, observed by Descloizeaux on the minute scales of the Pontivy mica, as sufficient ground for separating the new mineral from the old species. We should include under this species all hydrous micas which are rendered by the basic hydrogen orthosilicates, and to it the name Damourite belongs by priority. Sterlingite is simply a variety of Damourite, having the ratio 1:3:4, with more marked qualities and a wider optical angle than the Pontivy mineral; and, provisionally, the name I have given will be useful in designating it.

The hydrous micas, of which Sterlingite is a variety, have a special interest in connection with the subject of this paper, because they illustrate the characteristics of basic water, which will be contrasted, with those of water of crystallization, in our description of the following species. The evidences that the water in these micas is basic, — that is, forms a part of the basic radical, — may be summed up as follows:—

1. The amount of water in the different varieties is very variable, and bears no constant ratio to that of the other basic radicals.\*
2. The hydrogen of the water supplements the other basic radicals and fills out the amount required for a unisilicate, the type to which most of the micas conform.
3. The water is expelled only at a very high temperature.
4. The loss of water is not attended with any marked change in the appearance of the mineral.†

*Jefferisite*, of *West Chester*. — This well-known mineral, found in the serpentine at West Chester, Pa., was, as I have said, carefully analyzed by Professor Brush, of New Haven, who named it after W. W. Jefferis, Esq., of West Chester; and to this gentleman I am indebted for the specimens of the mineral whose crystallographic relations I have studied. As I am informed by this careful mineralogical observer, the locality is some three miles south of West Chester, where

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\* This fact is not shown so forcibly by the analysis cited above as by the series given by Professor Dana, on pages 310 and 311 of the fifth edition of his "System of Mineralogy," to which we would refer in illustration of this point.

† Since the above was written, we have received from Dr. F. A. Genth his very valuable paper on corundum and its associated minerals. He regards Damourite as one of the most important products of the alteration of corundum, and gives a large number of analyses of specimens from different localities, to which we gladly refer, as they illustrate the point made in this paper, even more markedly than the analyses cited above. The Damourites are evidently widely distributed minerals and characteristic features of certain rocks.

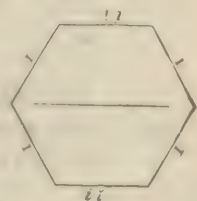


the mineral occurs in several veins or pockets of the serpentine, not more than fifty yards from the quarry where the ripidolite is found. It is associated with oligoclase; and corundum also occurs in the same serpentine formation, in Newton township, ten miles distant, on the north edge of the serpentine ridges. The Jefferisite is found in broad crystals, or crystalline plates, which frequently have a distinct hexagonal form. On two of these plates sent me by Mr. Jefferis, measuring respectively two inches and three-fourths of an inch in diameter, the form is as sharp as on crystals of Muscovite, and the edges make an angle of  $120^\circ$ , as nearly as it can be measured. Mr. Jefferis writes, — "In regard to the Jefferisite crystals which you have examined, the large crystals or plates occurred in the vein immediately under the sod, exposed to all frost, &c.; the small crystals were taken out four or five feet below, in the decomposed Deweylite, and were associated with striated oligoclase, of which I sent you specimens."

The crystals of Jefferisite cleave like mica, affording thin but unelastic foliæ. The cleavage planes are marked triangularly by lines, crossing at angles of  $60^\circ$  and  $120^\circ$ . In some cases there is a jointing as in crystals of mica, parallel to the shorter diagonal of the rhomb. One crystal sent me by Mr. Jefferis is the half of a rough hexagonal prism, an inch and a quarter high by two inches in diameter. The plane of the optical axes, as in the larger number of micas, is parallel to one of these lines, as indicated in Fig. 1, coinciding with the shorter diagonal of the rhombic prism, which appears to be the fundamental form in all this class of minerals, and from which the hexagonal form is derived by the truncation of the two acute angles. The double refraction is strongly negative, but the angle between the optical axes varies in the most remarkable manner. I have measured angles on different plates, of  $27^\circ$ ,  $24^\circ$ , and  $10^\circ$ , and observed many intermediate conditions. Owing to the deep yellow color, the plates become opaque at a very moderate thickness, and for this reason it is impossible to measure the angle with great precision. Some of the plates are apparently uniaxial; but this may result from the blending of the two hyperbolas, due to the thinness of the plate. The dispersion of the axes is but slight, and only perceptible in the thicker laminae when  $\rho < v$ . It is obvious, therefore, that the crystallographic characters of the mineral are identical with those of mica.

The plates are generally, if not invariably, twinned, and the twin-

Fig. 1.



ning is the cause of this most remarkable variation in the optical angle, as will be explained at length in connection with our description of Culsageeite. On this last mineral the same phenomena are more marked, owing chiefly to the greater transparency of the plates.

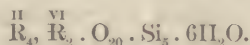
In order to illustrate the chemical relations of the mineral to the Biotite micas, we give below: 1st. The results of the analysis of Jefferisite by Professor Brush. 2d. The same results, calculated for the anhydrous mineral. 3d. The results of an analysis of a Biotite mica, from Pargas, Finland, by Svanberg. In each case I have added the amounts of oxygen in the several oxides, to show the atomic ratios:—

	Si	Al	Fe	Fe	Mg	Ca	K	H	
(1)	37.10	17.57	10.54	1.26	19.65	0.56	0.43	13.76	= 100.87
	19.78	8.18	3.16	.28	7.86	.16	.07	12.23	
	19.78	11.35			8.37			12.23	
	5.	3.			2.			3.	

	Si	Al	Fe	Fe	Mg	Ca	K		
(2)	42.94	20.33	12.20	1.46	22.75	.66	.50		= 100.84
	22.90	9.47	3.66	.32	9.10	.19	.08		
	22.90	13.13			9.69				
	5.	3.			2.				

	Si	Al	Fe	Mn	Mg	Ca	K	H	F	
(3)	42.58	21.68	10.39	.75	10.27	1.04	8.45	3.35	.51	= 99.01
	22.71	10.10	3.12	.17	4.11	.30	1.43	2.98		
	22.71	13.22			8.99					
	5.	3.			2.					

The general symbol of Jefferisite deduced from (1) would be,—



A comparison of the results given in (2) and (3) will show that the anhydrous Jefferisite corresponds very closely in its chemical constitution with the Biotite mica from Pargas. The chief difference is to be found in the fact that the mica contains potassium and basic hydrogen, in place of more than one-half of the magnesium of the Jefferisite. It should, however, be remembered in this connection that the Biotites present a very wide variation in the ratio between the amounts of the



protoxide and sesquioxide radicals which the various varieties contain. The limits usually assigned to this variation correspond to the ratios —

$$\overset{\text{II}}{\text{R}} : \overset{\text{VI}}{\text{R}} : \text{Si} = 1 : 2 : 3, \text{ and } \overset{\text{II}}{\text{R}} : \overset{\text{VI}}{\text{R}} : \text{Si} = 1 : 1 : 2,$$

and the Pargas mica with the ratio 2 : 3 : 5 falls between these limits ; but the Calsagee variety of vermiculite corresponds to the more common class of Biotites, which have the ratio 1 : 1 : 2.

But this resemblance in chemical constitution only appears when we compare the Biotite mica with the anhydrous Jeffersite ; while it is the crystallized hydrous Jeffersite which so closely resembles the magnesian micas in its crystallographic relations ; and the question now arises, What is the condition of the large amount of water —  $12\frac{1}{2}$  per cent — which the crystallized mineral contains ?

To aid us in forming a conclusion on this point, we have the following evidence :—

*First.* As the above analysis shows the water is united in definite and atomic proportions amounting to six molecules to every five molecules of silicon in the molecules of the mineral, that is sufficient to convert all the silicon into a hydrate, assuming that the five silicon atoms in this hydrate are joined to each other by the smallest possible number of bonds.

*Secondly.* While both the crystallographic and the chemical relations of Jeffersite to the other vermiculites, and to the magnesian micas, indicate that the mineral is an orthosilicate, the amount of basic radical, exclusive of the water, is amply sufficient to saturate the atomicity of the silicon.

*Thirdly.* It was observed by Professor Brush — and his observations have been fully confirmed by ourselves — that the water is given off at a comparatively low temperature, — about  $300^{\circ}$  C. : and, as every mineralogist knows, this dehydration is attended with that remarkable exfoliation which is characteristic of the vermiculites, and indicates a complete disintegration of the molecular structure. This exfoliation is wholly different from the phenomena which the so-called hydrous micas present under like conditions. In these last — which, as we suppose, contain hydrogen as a part of the basic radical of their molecules — a very high temperature is required to expel the water, and the loss is attended by no such marked change of volume and disintegration.

The conclusion that we draw from these facts is, that the combined water is in the same condition in Jeffersite as in the zeolites, and in many crystallized salts, — a condition which has long been known in chemistry as *water of crystallization*. We shall not here attempt to

discuss what are the relations of the water thus combined to the molecular structure of the mineral. This question is still in suspense, and we are persuaded that our science is not yet in a condition to solve the problem. All that we can at present do is to classify the phenomena presented by the exfoliation of the vermiculite minerals with the efflorescence of Glauber's salt or the intumescence of alum.

If, then, I am justified in the inferences I have drawn, Jefferisite differs chemically from Biotite mica chiefly in containing water of crystallization, very much in the same way that gypsum differs from anhydrite, or common salt from the crystals with two molecules of water, which form in brine at a low temperature. In these bodies, however, the forms of the crystals of the hydrous are wholly different from those of the anhydrous salts, while the crystallographic relations of Jefferisite are very similar, if not identical, with those of the magnesian micas. It is, however, also true that the crystalline structure of the micas seems to admit of a very wide variation of chemical composition. In cryophyllite, which has all the crystallographic characters of the micas strongly developed, is the atomic ratio 3:4:14; while in the Biotite, whose analysis was cited above, it is 2:3:5. The first is an acid, and the last a neutral silicate. The other varieties of mica have a composition intermediate between these extremes; and it might be said, if such a variation of composition is compatible with the crystalline structure exhibited by this group of minerals, it would not be surprising if the structure were sufficiently elastic to admit of the insertion of the water of crystallization without great alteration of external form.

It may be further urged that the normal mica crystal (a rhombic prism with angles of  $60^\circ$  and  $120^\circ$ ) is a form which appears to be related to a mode of molecular structure common to a great many substances, and the phenomena which will be described in this paper seem to sustain this opinion. Still, we do not care to theorize about the subject. Our object is simply to make prominent the two points: 1st. That the crystallographic structure of Jefferisite is identical with that of the magnesian micas. 2d. That the chemical constitution of the anhydrous mineral is closely allied to that of Biotite.

*Culsageeite or the Vermiculite of the Jenks Mine, North Carolina.*—Through the kindness of Colonel C. W. Jenks, the proprietor of the corundum mines on the Culsagee River in Macon County, N. C., I have had an opportunity of examining a variety of vermiculite which at that locality forms an important part of the matrix of the corundum. The associations of the mineral in North Carolina are very similar to those



in Pennsylvania. Professor J. Lawrence Smith, in his recent article in *Am. Jour. Sci.* (III. vi. 180), says that "in the development in North Carolina the corundum" (with which the vermiculite is associated) "occurs in chrysolite or serpentine rocks, and outside of serpentine it has not been found. These chrysolite rocks belong to a regular system of dikes which have been traversed for the distance of about one hundred and ninety miles. This system of dikes lies on the north-west side of the Blue Ridge, and has a strike parallel to the main mass of the ridge, and has an average distance from the summit of the ridge of about ten miles. The serpentine appears at intervals along this whole line of one hundred and ninety miles," and "is usually enclosed in a hard crystalline gneiss." In the serpentine the corundum has been found at several localities along a distance of forty miles. Colonel Jenks writes me: "The workmen have opened a new vein they call the gem vein. It is walled with chrysolite, and the fissure is from one to four feet wide, which fissure is filled with ripidolite, and in this the corundum crystals are imbedded." As regards the association of the ripidolite and vermiculite, he also writes: "They occur both alone and intermingled, and also in layers, like the leaves of a book." Again: "The ripidolite maintains a uniform character of texture and appearance, whether associated with the vermiculite or alone. In the vein at the bottom of the shaft I have spoke of, and elsewhere, it is of uniform hardness, texture, &c., but in it the corundum seems to have lost cohesion, — in some localities falling to pieces, and crumbling out of the ripidolite matrix. But, when exposed to the air and sunshine for a day or two, the corundum regains its cohesion, and you can then chip off the ripidolite from it. It is just the opposite with the vermiculite when alone the matrix. Sometimes it occurs in masses, several feet long, formed of scales as large as your hand, easily separating like mica from the corundum, which in this matrix always seems to retain its cohesion. Then, again, the vermiculite will occur in masses of many bushels, all broken up and disintegrated, like very coarse bran, in which the crystals of corundum lie like eggs in a box of sawdust. The great crystal (weighing 316 pounds) was thus deposited. At other places the entire mass seems to have changed to the color of white Castile soap, with specks of green in it, the corundum crystals all falling apart until exposed, when they harden." This untechnical language of an unprofessional but accurate observer conveys a very vivid idea of the mode of association of these minerals. The specimens so kindly sent me by Colonel Jenks confirmed in all respects his observations. In several cases the corundum was still imbedded in both the ripidolite and the

vermiculite, and the scales of these minerals not unfrequently penetrated the corundum crystals in such a way as to indicate that the corundum

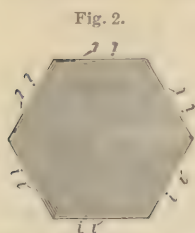


Fig. 2.

had crystallized around them. The ripidolite is firm in texture, and closely resembles the variety from West Chester. It occurs in hexagonal plates, which have a striation similar to that I formerly described as characteristic of the Pennsylvania specimens (*Am. Jour. Sci.*, II. xlv. 201, 1867), and which is illustrated by Fig. 2. Professor Smith, *loc. cit.*, gives analyses of two varieties of ripidolite—one compact, the

other friable—as follows:—

COMPACT RIPIDOLITE FROM NORTH CAROLINA.

Si	Al	Fe	Mg	H
27.00	21.60	16.63	22.00	12.30 = 99.53

FRIABLE RIPIDOLITE FROM NORTH CAROLINA.

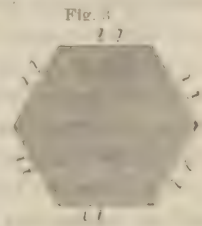
Si	Al	Fe	Mg	H
29.15	10.50	23.50	25.44	10.04 = 98.63

A determination of the silica in the compact mineral made by Mr. F. Gooch, student in the Laboratory of Harvard College, gave 27.25 per cent of  $SiO_2$ , and 11.93  $H_2O$ , as a mean of two closely agreeing determinations. But in the specimens sent me by Colonel Jenks I did not recognize the friable variety.

The vermiculite occurs in close contact with the ripidolite, and is frequently interlaminated with it, *but the two are always perfectly distinct*, thus entirely disproving the theory that the vermiculite was derived from the ripidolite by weathering. It occurs in large plates having more or less of an hexagonal outline. Some of those received from Colonel Jenks were five inches in diameter. It has a greenish-yellow color, which is very much lighter than that of the West Chester variety. The plates are strongly marked by lines crossing at angles of  $60^\circ$  and  $120^\circ$ , like those from West Chester; but these lines are more marked in the North Carolina variety. This variety is also much more friable than the other, and readily breaks in directions parallel to these lines,—yielding rhombic plates with angles of  $120^\circ$  and  $60^\circ$ , and more readily hexagonal or triangular plates, produced by the truncation of the  $60^\circ$  angle of the rhombic plate, on a line parallel to the shorter diagonal of the fundamental rhomb. The plates most readily break



parallel to this diagonal, and, like the specimens from West Chester, are frequently jointed in this direction. Like other micaceous minerals, the plates cleave readily parallel to the basal plane, yielding very thin foliae, exceedingly flexible, but not elastic. The optical characters are the same as those of the West Chester variety, — strong negative double refraction yielding a biaxial ring system, with uniform distribution of colors, and very variable optical angle. I have measured angles from about  $30^\circ$  to about  $13^\circ$ . The angle often varies widely in different parts of the same plate. Thus I have measured on different laminae, from a single plate not exceeding 3 inches in diameter, the three angles  $30^\circ$ ,  $24^\circ$ , and  $13^\circ$ ; and again I have noticed a similar variation on one and the same lamina. Indeed, the phenomena which I observed were almost identical with those I had previously observed on plates of ripidolite from Texas, Pennsylvania, *loc. cit.* On moving the lamina just referred to parallel to itself, in front of the polarizing microscope, the optical angle varied as I passed from one side of the field to the other. Beginning with a value of about  $30^\circ$ , the angle decreased to about  $13^\circ$ . Moving the plate still further, I found a region of indistinctness, and then the axes opened again, — the new plane making an angle of  $120^\circ$  to the old. I had evidently here a macle precisely similar to that I had previously described in the Texas ripidolite, and shown in Fig. 3, where the lines of shading represent the position of the plane of the optical axes. This represents what we may call an ideal macle; for I have seldom been able to trace more than three individuals on the same plate, and, as a rule, these are very unequally developed. On many of the specimens of the North Carolina vermiculite, the macle is externally marked by the eminent cleavage or jointing parallel to the shorter diagonal of the rhomb section, and in several of the specimens I have examined it was quite symmetrical. A study of these specimens led me to an explanation of the cause of the remarkable variation of the optical angle, which I believe not only applies to the vermiculites and ripidolites, but also, in many cases at least, to the micas. It would be expected that the several members of such macles as Fig. 3 represents would penetrate each other, and I therefore made a series of experiments to ascertain what would be the effect of the interfoliation of laminae in which the planes of the two sets of optical axes had the same relative position as in the several members of the macle. To that end I divided a plate, which presented the largest optical angle I had observed, into as thin laminae as possible, and then superimposed them in the relative



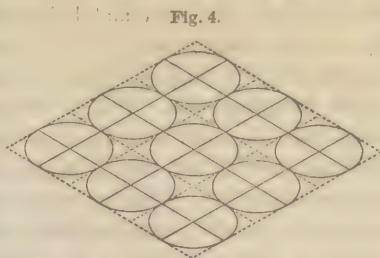
position I have mentioned, so that the planes of the optical axes should be inclined at an angle of  $60^\circ$ . The result was — when the thickness of the plates in each position were nearly equal — that a symmetrical ring system was obtained, in which the optical angle was about  $13^\circ$ , — the smallest I had measured; and, by varying the relative thickness, intermediate degrees of optical divergence were produced. By now introducing laminae into the compound crystal, in the position of the third member of the macle, — that is, with the plane of the optical axes in the position of the third diagonal of the hexagon, — the apparent angle could be reduced still further, so that the plate was apparently uniaxial. Although these experiments were sufficient to show that the macleing was an adequate explanation of the apparent variation of optical angle I had observed in the plates of ripidolite and Jefferisite, they also raised the question how far the effect I had obtained in my experiments might be due to the circumstance, that, on account of the deep color of these minerals, it is only possible to experiment on very thin plates, with which, of course, the rings of interference are very wide, and the hyperbolas proportionally indefinite. I therefore next made a similar experiment with a well-known phlogopite mica from Jefferson County, N. Y., whose crystals are very distinctly macle after the type of Fig. 3 or 5, the plates presenting a variation of optical angle similar to that I have described, the normal axial divergence being about  $15\frac{1}{2}^\circ$ . A very clear portion of one of these plates was first cut into a regular hexagon, one of whose diagonals was in the plane of the optical axes. This hexagonal plate was then split into twelve laminae, which were superimposed with the intervention of balsam, and in alternating positions, like the members of a macle, — the optical plane in each of the laminae making an angle of  $60^\circ$  with that of the lamina above or beneath it. The result was an essentially uniaxial plate, differing from a plate of uniaxial mica only in small irregularities in the contour of the rings, such as the lamination would be expected to produce. On repeating now this experiment with a Muscovite mica having a wide optical angle about  $63^\circ$ , I obtained a most remarkable and unexpected result, — a structure presenting optical phenomena similar to those of a plate of quartz cut perpendicularly to the principal axis. At the first trial I obtained a compound mica plate showing a disk of color in the centre of the field whose tint changed during the rotation of the analyzer, — of the polarizing microscope, — like a plate of left-handed quartz; and, on superposing a plate of right-handed quartz, the spirals of Airy at once appeared. The rings, however, were wholly broken up, and appeared only in irregular



patches of color. This irregularity was due to the unequal thickness of the laminae employed, and I found it impossible to split up one and the same hexagonal plate of mica into laminae, which were sufficiently uniform for the purpose. But very satisfactory results were obtained in the following way. I selected for the purpose the very clear and easily cleavable mica from Grafton, N. H.; and, after a few trials, succeeded in cleaving off very thin plates of considerable size and nearly uniform thickness. Selecting one of these plates, I first divided it by means of a parallel ruler into strips, and from these strips cut out the hexagonal laminae by means of a steel pattern carefully made. The thin mica can be cut with perfect accuracy by a sharp knife on a plate of glass. The hexagonal laminae thus obtained, though coming from different parts of the mica plate, were optically parallel to each other; and by drawing at the outset a line, with a sharp point, near the corresponding edges of the several strips, this line served as a guide for placing the hexagonal laminae. From laminae thus prepared, plates were made showing the familiar system of rings as perfectly as could be expected. The best results were obtained with plates consisting of from twelve to twenty-four laminae; and the character of the resulting plate, whether left or right handed, was found to depend on the order of the spiral arrangement. If in building up the pile the marked side of each successive lamina is turned through an angle of  $60^\circ$  in the direction of the motion of the hands of a watch, the result corresponds to left-handed quartz if turned in the reverse direction to right-handed, and on superposing two dissimilar plates thus prepared I obtained again the spirals of Airy in great perfection. Thus, then, it appears that, even with micas of the widest optical angle, we can build up a structure which is optically uniaxial.\*

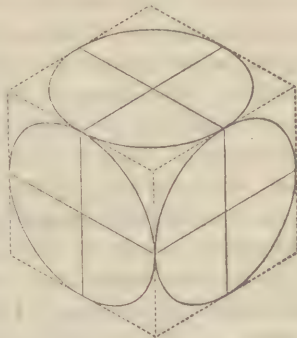
\* The great difficulty in preparing these plates is to obtain thin films of mica of uniform thickness, which are of sufficient size to yield a dozen or more laminae; and the more nearly we have succeeded in preparing such a film, by splitting sheets of mica, the more closely we have been able to imitate the phenomena seen under like conditions with a plate of quartz. We have been able to work with films which measured with a spherometer only  $\frac{1}{100}$  of an inch in thickness, and have not obtained good results with those which were much thicker, and when thinner than this the mica cannot readily be cut into shape. The least inequality in the thickness of the several laminae composing the same plate more or less mars the effect; and, although some of the striking features seen with quartz may remain, such as the succession of colors on revolving the analyzer, and even the spirals of Airy, yet the more delicate phases of the phenomena disappear. The plate changes color when revolved in its own plane, the rings lose their circular form and become confused, and the violet cross disappears. Moreover, as regards the conditions which determine the phase of the circular polarization, the law stated above can only be affirmed with cer-

The theory which I have formed to account for these facts is as follows. We may conceive that the molecule of mica is an ellipsoid, whose elliptical section, through the longer axis, can be inscribed in the rhomb of  $60^\circ$  and  $120^\circ$ . Assume now that these molecules have polarity, the rhombic prism would be the normal result of their association, as represented in Fig. 4. We



may, however, conceive that three of these molecules may become associated by alternate single poles to form a nucleus, such as represented in Fig. 5. Such a group once formed would be in a condition of great stability, resulting from the concurrent action of the several poles; and if now each of the molecules develops into a crystal, the result would be a macle of a form which is very common in the mineral kingdom. If six molecules unite in a similar way to form a nucleus, as in Fig. 6, we should also have stable equilibrium (although less firm than before), and the result of a symmetrical development would be a macle such as has been represented in Figs. 2 and 3. Whether this more complex arrangement is necessary in order to explain the phenomena presented by

Fig. 5.

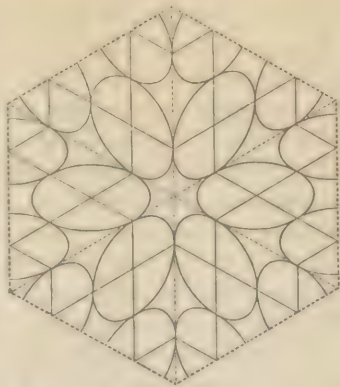


tainty of plates consisting of laminae which very nearly fulfil the conditions we have described. Very small inequalities of thickness renders the effect irregular, and made it at first difficult to discover the law. Our experiments have been a series of approximations; and, although we may never be able with our rude appliances to compete with nature in the manufacture of uniaxial crystals, yet we have approached so near to the perfect result as to be able to point out with confidence one way, at least, by which the effects seen in natural crystals may be produced. We have usually cut the laminae into regular hexagons, but equilateral triangles might more easily be cut, and would probably give as good results; for, although the errors of position might not be so well distributed, we have found that a slight variation in the relative position of the laminae injures the result to a far less degree than the least inequality in their thickness. It still remains to make careful quantitative measurements of the effects produced under determinate conditions; and, as can easily be seen, the subject opens a wide field for mathematical analysis as well as physical investigation.



the vermiculites and micas, I do not feel confident. It is seldom that more than three individuals can be distinguished on a given plate; and the very unequal development of the several individuals, and the indefiniteness of the lines of demarcation, resulting from the phenomena which have been described, render what would seem to be a characteristic feature of the more complex group not necessarily a certain indication of the structure. I refer to the fact, very constantly noticed, that the plane of the optical axes is parallel to the nearest hexagonal edge, as shown in Figs. 2 and 3. In Figs. 5 and 6 this same plane is parallel to the shorter axis of the ellipse; and it can easily be seen that if either of the individual of Fig. 5 were developed over any large portion of the space of its neighbors, the optical plane might appear parallel to the adjacent edge.

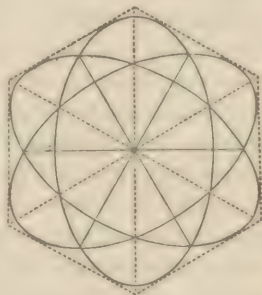
Fig. 6.



Having made the two suppositions, as above, to explain the phenomena of twinning, which have been long familiar and externally visible, it will not, we trust, appear unreasonable if we make a third supposition to explain the phenomena first described in this paper. We may conceive that the ellipsoidal molecules, instead of grouping together on the same plane, become associated by their alternate poles, one over the other, as represented in Fig. 7.

Molecules so associated, developing laterally, would produce the laminæ of a mica plate in the relative position in which we have placed them in our artificial crystals, with only this difference, that the laminæ would be indefinitely thin, and in exact position; and the effect of such compound molecules in modifying the elasticity of the crystalline structure must be, in most respects at least, like that of single molecules, symmetrical on all sides of one line

Fig. 7.



or axis.—in other words, they must produce a structure similar to that of uniaxial crystals. Under what further conditions the grouping of

the molecules, in right or left handed spirals, determines the phenomena of right or left handed circular polarization, and what bearing the new facts may have on the received theory of these phenomena as they appear in quartz, must be left for further analysis to discuss.

I pass next to consider the composition of the Culsagee vermiculite, and I give below, at (1), (2), and (3), the results of three analyses, made by myself, together with the corresponding oxygen ratios.

The pulverized mineral, after it has been exfoliated by heat, is easily and perfectly decomposed by hydrochloric acid. In analysis (1), after the separation of the silica, the alumina and ferric oxide were separated from the magnesia by ammonia, with the usual precautions. In (2) and (3) the bases were converted into nitrates and separated by Deville's method. In each case the magnesia was weighed as pyrophosphate, and the alumina and ferric oxide were weighed together.

All three analyses were made with material rendered anhydrous by ignition until the weight was constant, and each is represented by three distinct determinations; namely, the weight of the silica, the sum of the weight of the alumina and ferric oxide, and the weight of the magnesian pyrophosphate. The oxides of iron and the water were once for all determined on separate portions of the dried but not exfoliated mineral, and therefore appear of the same value in all the analyses.

The determination of the water was the only difficulty which the analysis of this mineral presented. It is by far the most hygroscopic silicate I have ever examined, when once dry absorbing water from comparatively dry air with almost as much avidity as chloride of calcium. In two experiments with different portions of the same powder, the material was heated in an air bath, at  $100^{\circ}$  C., for seventy-two hours before the weight became constant; and in each case the weight was compared at intervals of about six hours. The total loss in the first experiment was 10.27 per cent, and in the second 10.19 per cent. The mineral thus dried lost when ignited 10.84 per cent. Another portion of the same powder, which had been dried over sulphuric acid for more than two months, lost when ignited 11.09 per cent. This close agreement indicates that all the water lost in drying, either at  $100^{\circ}$  or over sulphuric acid, is hygroscopic; and the conclusion is confirmed very greatly by the fact that the mineral in thus drying does not change its aspect in the least degree, and rapidly reabsorbs the water when exposed to the air. On the other hand, when the mineral is ignited it swells up to many times its volume, like other members of the vermiculite family, and undergoes what is evidently a profound alteration in its molecular structure.\*

\* In his valuable paper on corundum, above referred to (p. 44), Dr. Genth



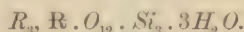
	Si	Al	Fe	Fe	Mg	H
(1)	37.58	19.73	5.95	0.58	25.13	11.09 = 100.06
	20.04	9.19	1.78	0.14	10.05	9.86
	20.04	10.97		10.19		9.86
	2.03	1.11		1.03		1.
(2)	37.43	19.75	5.95	0.58	25.58	11.09 = 100.38
	19.96	9.20	1.78	0.14	10.23	9.86
	19.96	10.98		10.37		9.86
	2.03	1.11		1.05		1.
(3)	37.10	20.22	5.95	0.58	25.07	11.09 = 100.01
	19.79	9.42	1.78	0.14	10.03	9.86
	19.79	11.20		10.17		9.86
	2.	1.13		1.03		1.

has published two analyses of the Calsagee vermiculite, made in his laboratory by Mr. Chatard and Dr. Koenig. Their results agree very closely with those given above, except in the amount of water; and since, as Dr. Genth states, the analyses were made with material which had been dried *in vacuo* over sulphuric acid for only four days, it is obvious that they have estimated as combined water a large portion of what we, for reasons stated, have regarded as hygroscopic moisture. Reducing their results to the same standard as our own, the close agreement becomes evident. We give, for comparison, first the results, as published; second, the same reduced to a water percentage of 11%; third, the corresponding oxygen or atomic ratios.

	Si	Al	Fe	Fe	Ni	Mg	H
Mr. CHATARD.	33.93	17.38	5.42	0.50	0.35	23.43	19.17 = 100.18
	37.33	19.12	5.94	0.55	0.38	25.77	11.09
	19.91	8.91	1.78	0.12	0.07	10.31	9.86
	19.91	10.69		10.50			9.86
	2.02	1.06		1.05			1.
	33.77	17.56	5.61	0.50		22.48	20.30 = 100.22
Dr. KOENIG.	37.65	19.58	6.26	0.56		25.06	11.09
	20.08	9.12	1.88	0.12		10.02	9.86
	20.08	11.00		10.14			9.86
	2.04	1.11		1.03			1.

The close agreement of these results with our own, obtained by different processes and with different specimens of the mineral, indicates a remarkable constancy of composition for a micaceous mineral. I have confirmed Dr. Genth's observation of the presence of a very small amount of nickel in the mineral, and I also detected in it a trace of copper.

It is evident from these analyses that the atomic ratio of the mineral is 2 : 1 : 1 : 1, and its formula may therefore be written :—



By referring to what has already been said of the relation of Jefferisite to the Biotites, it will be seen that, while that mineral corresponds to a less common variety of this species, the Culsagee vermiculite corresponds to its more usual type. We give below, at (1), the results of an analysis of Vesuvian Biotite, by Chodnef; and, at (2), the same, assuming that a portion of the iron is in the ferrous condition, as is well known to be the case, in order to show that values within the probable error of the analysis would give the ratio 2 : 1 : 1. Here, of course, the alkali takes the place of a portion of the magnesia of the vermiculite.

	Si	Al	Fe	Fe	Mg	Ca	K
(1)	40.91	17.19	11.03		19.04	0.30	9.96 = 98.43
(2)	40.91	17.19	7.03	4.00	19.04	0.30	9.96 = 98.43
	21.82	8.30	2.11	.89	7.62	0.09	1.69
	21.82	10.41			10.29		
	2.12	1.01			1.		

This new variety of vermiculite is so well marked, and the composition so definite, that I have thought best to designate it by the name Culsageeite. As regards its other characters, it has a specific gravity of 2.225 (taken in alcohol), and about the hardness of talc. Before the blowpipe the exfoliated mineral fuses readily to a white enamel, but does not fuse in the flame of a Bunsen lamp.

To this variety of vermiculite belongs, as I have already intimated, the original mineral from Millbury, analyzed by Crossley. There can be no question as to the general accuracy of Crossley's results; and, assuming that all the iron is in the ferric condition, they give, as I have shown, almost precisely the atomic ratio 2 : 1 : 1 : 1. The only question that can arise is in regard to the condition of the iron. I have therefore made an assay of the iron by the accurate method I formerly described in the *Am. Jour. Sci.*, xlv. 347, 1867. One hundred parts of the massive mineral gave 7.40 per cent ferric oxide, and 3.86 per cent ferrous oxide, which corresponds to a total of 10.56 ferrous oxide. Crossley found 10.02 ferrous oxide, but he separated with great care the vermiculite from the steatite with which it is mixed; and this steatite was probably the source of the greater part of the small quantity of ferrous oxide found in our assay, which, though not made with



pure material, shows conclusively that the condition of the iron in the Millbury vermiculite is not different from that in other varieties of the same family of minerals.

*Hallite.* — Several years since this variety of vermiculite was sent to me by Mr. John Hall, of Philadelphia, by whom it was originally discovered. The examination then made showing that the mineral was a new variety, if not a new species, of the vermiculite family, I gave to it the name of Hallite, in recognition of the mineralogical services of Mr. Hall, who not only discovered the mineral, but has carefully worked the locality and observed the associations in which it is there found. A preliminary notice of the mineral under this name was published at the time by Professor Leeds, of Hoboken; but the interesting relations which the mineral bears to the subject of this paper have made a further examination desirable.

Hallite occurs in large rough six-sided prisms, with easy micaceous cleavage. There are two varieties, differing markedly in color, green and yellow; and I am indebted to Mr. Hall for the following facts in regard to the locality and associations of this species. Mr. Hall writes: "The mineral is found at East Nottingham, in the serpentine formation of south-eastern Pennsylvania, three miles south of Oxford, in Chester County; and I know of no other locality. I think the green and yellow varieties are very closely related, and may possibly pass from one into the other; but I have no positive proof that they do. The crystals are found in nests or pockets, and the two colors are not found in the same nests. The green crystals are imbedded in a steatite earth or base of the same color as the crystals, and the yellow in a yellow earth; and sometimes nests, containing the opposite varieties, are only a few feet apart in seams of the serpentine rock."

As the following analyses show, the two varieties have essentially the same composition, and the only difference that could be detected was in the degree of oxidation of the iron. The yellow crystals appeared to be more weathered than the green, and on the last the green color frequently fades out towards the centre of crystals, thus giving indications of a metamorphosis by which one variety may pass into the other.

Under the microscope these scales of the mineral show a remarkable appearance. Between the greenish or nearly colorless plates are seen elongated scales of a yellow mineral resembling closely in color thin scales of Jefferisite. They are more or less spear-shaped in form, although usually very narrow, and lie accurately in parallel lines, which cross at angles of  $60^\circ$  and  $120^\circ$ , like the magnetic oxide of iron in the Muscovite from Pennsbury, Pa., or the microscopic crystals in the

Biotite of South Burgess of Canada; and the phenomenon of asterism, seen so beautiful with the plates of the last, can also be seen with thin laminae of Hallite. It was impossible to free the mineral from this admixture, but specimens were selected for analysis as free from it as possible. It was also impossible to determine its exact nature. The scales had not a definite form, but there was a tendency to a rhombic shape, which is well described by the term "spear-shaped;" and though the material is so widely distributed through the crystal, the total mass must be very small.

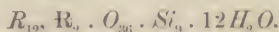
This mineral is not so hygroscopic as Jefferisite, and no difficulty was found in drying the material for analysis. When ignited, it exfoliates like other species of vermiculite, but not nearly to so great an extent as Jefferisite. After ignition it is decomposed by hydrochloric acid. The specific gravity of the green variety, mean of four determinations, 2.398; that of the yellow variety, mean of two determinations, 2.402. Before the blowpipe fuses with difficulty to a brown enamel. The following analyses were made by Mr. C. E. Munroe, Assistant in the Laboratory of Harvard College:—

## GREEN VARIETY OF HALLITE.

	Si	Al	Fe	Fe	Mg	K	H
(1)	35.97	7.61	8.83	1.13	31.34	0.43	14.32 = 99.63
(2)	35.80	7.29	8.73	1.13	31.56	0.49	14.33 = 99.33
(Mean.)	35.89	7.45	8.78	1.13	31.45	0.46	14.33 = 99.49
	19.14	3.47	2.63	0.25	12.58	0.08	12.74
	19.14	6.10		12.91			12.74
	3.	0.96		2.02			2.
	3.	1.		2.			2.

## YELLOW VARIETY OF HALLITE.

	Si	Al	Fe	Fe	Mg	K	H
(1)	35.17	7.74	9.76	0.32	31.61	0.56	14.65 = 99.81
(2)	35.34	7.42	9.61	0.32	31.41	0.65	14.91 = 99.66
(Mean.)	35.26	7.58	9.68	0.32	31.51	0.61	14.78 = 99.74*
	18.81	3.53	2.90	0.10	12.60	0.10	13.14
	18.81	6.43		12.80			13.14
	3.	1.03		2.04			2.09
	3.	1.		2.			2.



\* Trace of manganese.

It will be seen from the above analyses that, although the atomic ratio between all the basic radicals and the silicon is the same as in Calsagite, Jefferisite, and Biotite, the ratio between the protoxide and sesquioxide radicals is very different. In this respect the mineral resembles the phlogopite micas, in which also the protoxide radicals preponderate; and the symbol given above for Hallite, less the water, is identical with that given by Professor Dana as the more probable formula of the phlogopites.

The opacity produced by the interspersed material made it difficult to determine the optical characters of the mineral, as the rings produced with polarized light could only be seen with very thin plates, and the cross was therefore ill defined; so that, although in some cases there appeared to be a separation of the hyperbolas, the plates could not be distinguished from uniaxial. On one specimen the hexagonal form was very perfect, and the crystal presented the planes of a rhombohedron having an angle over the basal edge of about  $122^\circ$ , resembling the crystals of Biotite from Greenwood Furnace. Mr. Hall informs me that these more perfect crystals have only been found in one pocket of the serpentine.

The distinction, however, between the phlogopites and the biotites is not fundamental, either chemically or physically. Chemically, both species are orthosilicates; that is, the atomic ratio between the silicon and the sum of the basic radicals is 1 : 1. The species differ in composition only in the relative proportion of the sesquioxide and protoxide radicals. In the phlogopite the ratio of  $\overset{\text{II}}{R}$  to  $\overset{\text{VI}}{R}$  is probably normally 2 : 1; but of the published analyses the value varies between that ratio and the ratio 3 : 2. In the biotites the same ratio is probably normally 1 : 1; but here, again, the different analyses which have been made give values varying between 5 : 3 and 1 : 2. In like manner the optical distinction between the phlogopites and biotites, of which so much has been made, is equally indefinite. Between a so-called phlogopite, like that from Jefferson County, N. Y., with an angle of about  $15^\circ$ , and the apparently uniaxial plates of biotite from Vesuvius, there is every possible gradation — sometimes, as I have shown, on one and the same mica plate; and I have endeavored in this paper to explain the cause of this variation. With the Vesuvian biotites themselves, — if the specimens in the mineralogical cabinet of Harvard College are fair representatives of the mineral from this locality, — it is only occasionally that we find a perfectly uniaxial plate. More commonly there are distinct evidences of twinning, and on the borders of the hexagonal plate may be dis-



covered a biaxial structure of which the optical plane is parallel to different edges of the hexagon on different parts of the plate.

It must, however, be remembered that, as by the process of twinning we have described the structure of the magnesian micas approaches that of *biaxial* crystals, rhombohedral and other planes characteristic of the hexagonal system begin to appear on the crystal. This is illustrated not only by the crystals of Biotite from Vesuvius and from Greenwood Furnace, N. Y., but also by the more perfect crystals of Hallite from Chester County, Pa. In other words, *the process of twinning we have illustrated in this paper produces hexagonal crystals in external form as well as in optical characters*; and the question naturally arises, May not the hexagonal crystals of other minerals be formed in a similar way? — that is, may they not be developed from twinned molecules, which, though in their aggregate producing an hexagonal structure, singly would develop into biaxial crystals? Bearing on this point, we have discovered some very remarkable evidence.

We have in our possession a plate of Elba tourmaline cut perpendicular to the axis, in which the polarizing microscope shows on different zones a separation of the hyperbolas, which amounts in some positions to eight degrees; and in moving the plate across the field the optical divergence varies precisely as on plates of phlogopite and vermiculite. There is certainly no external evidence of lamination on tourmaline crystals, for the mineral is remarkably compact, and the crystals have not even a basal cleavage: but it will be remembered how readily some of the varieties pass by alteration into micas of the magnesian type; and this change to a foliated structure, in which the lamination is parallel to the base of the original hexagonal crystal, may be facilitated by a grouping of the molecules of the tourmaline, in the manner represented by Fig. 7.

We have also a plate of amethystine quartz, in which a beam of parallel polarized rays of light exhibits a twinning almost as symmetrical as that shown in Fig. 5, — the three zones being most beautifully mapped out by the alternating bands of right and left handed quartz, which are such a familiar phenomenon of these crystals; but, besides this, in each of these zones, near the border of the plate, can be distinguished a biaxial structure with an optical divergence of several degrees; and, on one other plate of amethyst we have had an opportunity of examining, we have also seen under the polarizing microscope the biaxial curves at one or more points.

These facts most distinctly suggest the theory that the optical phenomena of quartz are produced by a molecular structure similar to that

by which we have obtained identical phenomena in our artificial plates of mica, and that the two orders of crystals are aggregates of compound molecules, whose parts are twinned together in the one case in right-handed, and in the other in left-handed spirals, and, lastly, that the simple molecule, if developed normally, would produce a biaxial structure.\* This theory is most markedly in harmony with the chemical relations of silica. The compound  $SiO_2$  is the only one of the tetrad oxides which crystallizes in the hexagonal system; and ever since, by the study of the organic compounds of silicon, the quadrivalent character of the element has been made evident, this fact has been a striking anomaly in our chemical classification. Assume, however, that the molecule  $SiO_2$  would develop normally into a rhombic structure, and that the hexagonal form of quartz is solely a result of molecular twinning, and the anomaly disappears. The molecule  $SiO_2$  may be approximately of the same form as the molecule  $TiO_2$ , in Brookite; but, having the exact dimensions and polar conditions which favor the mode of molecular twinning, described above and represented by Fig. 7, it may always develop into hexagonal shapes.

Are, then, all hexagonal forms thus closely related to the rhombic systems of crystals? And do all molecules of the dimensions and polar conditions illustrated by the figures of this article—that is, those which correspond to the rhomb of  $60^\circ$  and  $120^\circ$ —usually develop into hexagonal forms? May not the whole difference between an hexagonal and a rhombic form arise from a slight difference of dimensions, which determines a molecular mauling in the one case, and a normal development of the single molecules in the other? These questions point out most interesting lines of investigation, and will recall to the mineralogist a number of facts bearing upon the subject. Allow me to refer to two of the most striking and most obvious.

On the crystals of chrysoberyl, the rhombic angle is  $119^\circ 46'$ ; and every mineralogist is familiar with the hexagonal mauling, similar

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\* Since the above was in type, we have received *Am. Jour. Sci.*, IV., February, 1874, containing a description of the rhombic silica which Professor Maskelyne, of the British Museum, has discovered in the meteorite of Breitenbach. This new species of silica, which Professor Maskelyne calls *Asmanite*, has the form of a right rhombic prism, with an angle of  $120^\circ 20'$ , and the crystals are optically biaxial; but while the specific gravity of quartz is 2.6, that of *Asmanite* is said to be 2.245. It is perhaps to be expected that such a molecular mauling as we have described would determine an increase of density, since thereby three molecules coalesce to form one; or it is possible that the remarks made in regard to calcite beyond apply also to quartz; but still the marked difference remains to be explained.

to Fig. 2, which is so very characteristic of this species.\* Corundum differs chemically from chrysoberyl, in that a portion of the alumina in the former is replaced by glucina in the latter. Corundum has a perfect hexagonal form, and, fundamentally, may not the only crystallographic difference be that, in consequence of the replacement, a rhomb of  $120^\circ$  changes to a rhomb of  $119^\circ 46'$ ? Now we have a plate of macle chrysoberyl, showing the normal wide divergence of the optical axes at certain points on its borders, and a nearly uniaxial structure at the centre, where there is an obvious interpenetration between the individuals of the macle, and where the superposition of the several laminae is most beautifully shown by a polarized beam of parallel rays. We have also a section of a corundum crystal, presenting phenomena similar to those seen with the plate of tourmaline, described above. Further, we have observed like phenomena on a section of phenacite; and, although the last mineral contains silica, yet if the molecules of  $SiO_2$  are crystallographically equivalent to those of  $Al_2O_3$ , it may be that the molecular structure both of phenacite and of beryl is more closely allied to that of chrysoberyl and corundum than the received theory of their chemical constitution would indicate.

We would not convey the impression that in all these crystals the appearances we have described are strongly marked, or that they have passed wholly unnoticed hitherto. Every one who has become familiar with the optical properties of crystals must have noticed that, with many always regarded as uniaxial, there is not unfrequently in some positions a small separation of the cross into the hyperbolas, which are characteristic of biaxial structure. But these irregularities, although long known, have never been satisfactorily explained. They have been hitherto residual features not accounted for by the received theory of crystalline structure, which explains so satisfactorily the general order of the phenomena observed with the polariscope. We have endeavored in this paper to trace their true significance: first, by showing that the appearances we are discussing are precisely similar to the effects which can be obtained by known means with mica plates; and, secondly, by observing on different specimens of various minerals every intermediate stage between the unmistakable effects of twinning on plates of mica or vermiculite, and the delicate phases of the same phenomena, seen with sections of crystals of tourmaline, corundum, or phenacite. One other illustration of our theory.

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\* See also Dana's System of Min., 5th ed., Figs. 154, 155, p. 156.



The rhombic angle of witherite (native baric carbonate) is  $118^{\circ} 30'$ , and the all but universal hexagonal macing of crystals of this species is a well-known fact.\* The rhombic angle of aragonite (the corresponding form of calcic carbonate) is  $116^{\circ} 40'$ , and the much greater divergence of this angle from  $120^{\circ}$  determines, as is also known, a style of macing which is usually quite different from that of witherite. In the isomeric calcite, however, we have the type of all hexagonal forms. Hitherto the crystalline forms of calcite and aragonite have been regarded as being as widely separated as possible, and a comparison of these two well-known mineral species has furnished one of the most striking illustrations of demorphism. But may not, after all, the comparatively small physical differences between these two minerals correspond to a crystallographic difference no greater, fundamentally, than the difference between the rhomb of  $116^{\circ} 10'$  and the rhomb of  $120^{\circ}$ ?

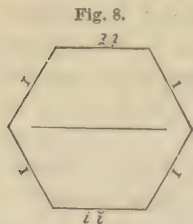
The macles of chrysoberyl and witherite are illustration of a general truth, fully recognized in mineralogy, that all rhombic crystals, whose angles approach  $120^{\circ}$ , tend to form hexagonal macles. The optical phenomena described in this paper certainly suggest the theory that a perfect hexagonal form and structure may be the result of a more fundamental and molecular macing, which results when the angle is exactly  $120^{\circ}$ .

*Observations of Senarmont.*—The only previous observations which we have been able to find bearing on the subject of this paper are those of the late eminent mineralogist, H. de Senarmont, of Paris. In a well-known paper (Ann. Chim. et Phys. 3<sup>e</sup> xxxiii. 391), Senarmont showed that salts which were both geometrically and chemically isomorphous might have very different optical relations; for example, that, while the biaxial crystals of two such salts might have the same bisectrix, the plane of the optical axes in the crystals of one might be perpendicular to the corresponding plane in those of the other. He further proved, by crystallizing together two salts so related, that in the crystals of the isomorphous mixtures thus obtained the optical angle varied with the varying proportions of the constituents, between the extreme conditions in the crystals of either salt; and by trial he succeeded in forming from two biaxial salts crystals which, in monochromatic light at least, appeared uniaxial. In a later paper (Ann. Chim. et Phys. 3<sup>e</sup> xxxiv. 171), Senarmont applied the principles, which he had thus experimentally verified, to explain the variation of the optical angle of the micas. In this paper he seeks to prove, first,

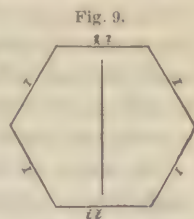
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\* See figures, Dana's System of Min., p. 697.

that all micas may be referred to a right rhombic prism with angles of approximately  $60^\circ$  and  $120^\circ$ ; secondly, that, while in some micas the plane of the optical axes is parallel to the shorter diagonal of the rhomb, as in Fig. 8, in others it is parallel to the longer diagonal, as in Fig. 9. Interpreting these facts by the results of his experiments on isomorphous salts, he draws the inference that there are, crystallographically at least, but two species of mica which are



geometrically isomorphous but optically distinct, that these are represented by the varieties which have the widest optical angle between the axes in either plane, and that all other varieties with optical angles varying from  $0^\circ$  to  $70^\circ$  in either direction are isomorphous mixtures of the two optically distinct conditions of the mineral.



The observations described in this paper, although they prove that another cause may also determine the variation of optical angle in micaceous minerals, do not necessarily invalidate this beautiful theory of Senarmont. The variations observed with other minerals, not only on different specimens, but with the same specimen at different temperatures, and which are so beautiful seen with the orthoclase from Wehr in the Eifel, and with crystals of selenite, indicate that such variations may be determined by conditions of molecular structure wholly independent of the macting here described. We have shown that the macting does produce the variation in certain cases, and it must remain for future investigation to assign the limits of the influence which this cause may exert. We would only remark in conclusion that, although in the 57 varieties of micas examined by Senarmont he did not note a single instance in which the position of the plane of the optical axes — with reference to the diagonal of the rhomb — was different on different parts of the same specimen, or even on different specimens from the same locality, he does describe and figure several remarkable mactes of muscovite mica similar to those of vermiculite described above and represented by Fig. 5. In one instance the plane of the optical axes is parallel to the shorter, and in the other to the longer diagonal of the rhombic prism; but in both cases it has the same relative position in the several individuals of the mactes, the three planes forming with each other angles of  $60^\circ$ . On the plate of another macle a difference of optical angle was observed on different portions of the plate, and this effect was probably similar to that we

have been studying in this paper. On another plate, from the same crystals he observed a superposition of the laminæ of the different individuals of the macle; and the following language by which this phenomena is described, and which is the only reference made to it, shows how closely he came to the results recorded in this paper:—

“Une partie seulement du cristal maclé est commune aux trois lames; mais ici certaines plages (pointillées sur la figure) ont un caractère optique tout particulier; elles ne cessent jamais de développer des couleurs dans la lumière polarisée, quelle que soit d'ailleurs l'orientation de la lame cristallisée. Il est évident que dans ces plages il y a superposition de lames appartenant à des cristaux orientés, les uns comme le petit, les autres comme le grand cristal, de façon que quand les unes ont leurs sections principales dans le plan de polarisation, les sections principales des autres font avec ce plan des angles de 60 degrés.

“Cet exemple prouve donc, non-seulement que les micas peuvent se grouper latéralement sans que leurs clivages cessent d'être parallèles, mais que des feuillets superposés peuvent même appartenir à des cristaux dont l'orientation diffère de 60 degrés. Un pareil mode de groupement, qui ne trouble ni la régularité de la cristallisation, ni la transparence, semble indiquer que l'arrangement moléculaire de ces prismes rhomboïdaux diffère très-peu de celui qui conviendrait au prisme hexagonal régulier.”







